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Carrier based on granules produced from pyrogenically prepared silicon dioxides

The present invention relates to the use of granules of pyrogenic silica as carriers. In addition to various other actions, the granules can have the function of a carrier for foodstuffs additives, such as dyestuffs, antioxidants, preservatives, emulsifiers, gelling agents, thickeners and binders, stabilizers, alkalis, acids, salts, antilumping agents, flavour intensifiers, sweeteners, aromas, feedstuffs additives, chemical intermediates and plant protection agents, such as, for example, herbicides, insecticides, fungicides and others.

It is known to employ spherical silicon dioxide particles as carriers, for example for feedstuffs additives (Sipernat 22, Bulletin Pigments No. 31, "Synthetic Silica as a Flow Acid and Carrier Substance", Degussa AG).

Disadvantages of the abovementioned silicon dioxide particles which are employed as carriers are their high water content, their too low purity and the poor flow properties of the loaded substance. Silicic acid esters, silica sols or silicates are employed as starting compounds, and then often lead to products of which the purity is not adequate for the desired intended uses because of considerable amounts of salts, so that an expensive washing is necessary.

The invention is therefore based on the object of providing spherical silicon dioxide particles for use as carriers

which do not have the disadvantages mentioned and moreover meet the high demands of uses in respect of purity, product safety and flow properties.

The invention provides the use of granules based on pyrogenically prepared silicon dioxide as a carrier for substances chosen from the group consisting of foodstuffs additives, such as dyestuffs, antioxidants, preservatives, emulsifiers, gelling agents, thickeners and binders, stabilizers, alkalis, acids, salts, antilumping agents, flavour intensifiers, sweeteners, aromas, feedstuffs additives, chemical intermediates and plant protection agents, such as herbicides, insecticides, fungicides and others.

The invention also provides an adsorbate of granules based on pyrogenically prepared silicon dioxide and at least one substance chosen from the group consisting of foodstuffs additives, such as dyestuffs, antioxidants, preservatives, emulsifiers, gelling agents, thickeners and binders, stabilizers, alkalis, acids, salts, antilumping agents, flavour intensifiers, sweeteners, aromas, feedstuffs additives, chemical intermediates and plant protection agents, such as herbicides, insecticides and fungicides.

The granules based on pyrogenically prepared silicon dioxide preferably have an average particle diameter of 10 to 120 μm and a BET surface area of 40 to 400 m^2/g (determination in accordance with DIN 66 131 with nitrogen).

The silicon dioxide granules furthermore preferably have the following physico-chemical characteristic data, which are determined as described in EP PS 0 725 037:

Pore volume:

0.5 to 2.5 ml/g

Pore size distribution: less than 5% of the total pore volume has a pore diameter of less than 5 nm, remainder meso- and macropores

:Hq

3.6 to 8.5

Tamped density:

220 to 700 g/l.

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Granules which are suitable for the use according to the invention and the preparation thereof are described, for example, in EP OS 0 727 037.

The granules can preferably have meso- and macropores, the volume of the mesopores making up 10 to 80% of the total volume. The particle size distribution of the granules is preferably 80 vol.% larger than 8 µm and 80 vol.% smaller than 96 µm. In a preferred embodiment of the invention, the content of pores smaller than 5 µm is not more than 5%, based on the total pore volume.

The granules employed according to the invention can be prepared, for example, by dispersing pyrogenically prepared silicon dioxide, preferably silicon dioxide prepared from silicon tetrachloride by means of flame hydrolysis, in water, spray drying the dispersion and optionally then heat-treating the resulting granules at a temperature of 150 to 1,100°C for a period of 1 to 8 h.

The dispersion in water preferably has a concentration of silicon dioxide of 5 to 25 wt.%, more preferably 5 to about 19.9 wt.%. The spray drying can be carried out at a temperature of 200 to 600°C, and disc atomizers or nozzle atomizers can be employed in this context. The heat treatment of the granules can be carried out either in a static bed, such as, for example, in chamber ovens, or in an agitated bed, such as, for example, rotary tubular dryers.

The pyrogenic silicon dioxide serving as the starting compound is prepared by a process in which a volatile silicon compound is injected into an oxyhydrogen gas flame of hydrogen and air. Silicon tetrachloride is used in most cases. This substance hydrolyses to silicon dioxide and hydrochloric acid under the influence of the water formed during the oxyhydrogen gas reaction. After leaving the flame the silicon dioxide enters into a so-called

coagulation zone, in which the silicon dioxide primary particles and primary aggregates agglomerate. The product present as a type of aerosol in this stage is separated from the gaseous concomitant substances in cyclones and then after-treated with damp hot air. The residual hydrochloric acid content can be lowered to below 0.025% by this process.

The granules based on pyrogenically prepared silicon dioxide can be silanized. The carbon content of the granules is then preferably 0.3 to 15.0 wt.%.

Halogenosilanes, alkoxysilanes, silazanes and/or siloxanes can be employed for the silanization.

The following substances can be employed in particular as halogenosilanes:

15 Halogeno-organosilanes of the type $X_3Si(C_nH_{2n+1})$

X = C1, Br

n = 1 - 20

Halogeno-organosilanes of the type $X_2(R')Si(C_nH_{2n+1})$

x = C1, Br

R' = alkyl =

n = 1 - 20

Halogeno-organosilanes of the type $X(R')_2Si(C_nH_{2n+1})$

X = Cl, Br

R' = alkyl

n = 1 - 20

Halogeno-organosilanes of the type $X_3Si(CH_2)_m-R'$

Halogeno-organosilanes of the type (R)X2Si(CH2)m-R'

Halogeno-organosilanes of the type (R)2X Si(CH2)m-R'

$$X = Cl, Br$$

$$R = alkyl$$

$$m = 0.1 - 20$$

$$R' = alkyl, aryl (e.g. -C6H5)$$

$$-C4F9, -OCF2-CHF-CF3, -C6F13, -O-CF2-CHF2$$

$$-NH2, -N3, -SCN, -CH=CH2,$$

$$-OOC(CH3)C = CH2$$

$$-OCH2-CH(O)CH2$$

$$-NH-CO-N-CO-(CH2)5-7$$

The following substances can be employed in particular as alkoxysilanes:

organosilanes of the type $(RO)_3Si(C_nH_{2n+1})$

R = alkyl

n = 1 - 20

Organosilanes of the type $R'_{x}(RO)_{y}Si(C_{n}H_{2n+1})$

$$R = alkyl$$

$$R' = alkyl$$

$$n = 1 - 20$$

$$x+y = 3$$

$$x = 1,2$$

$$y = 1,2$$

Organosilanes of the type (RO)3Si(CH2)m-R'

$$R = alkyl$$

$$m = 0,1 - 20$$

$$R' = alkyl, aryl (e.g. -C_6H_5)$$

$$-C_4F_9, OCF_2-CHF-CF_3, -C_6F_{13}, -O-CF_2-CHF_2$$

$$-NH_2, -N_3, -SCN, -CH=CH_2,$$

$$-OOC(CH_3)C = CH_2$$

$$-OCH_2-CH(O)CH_2$$

$$-NH-CO-N-CO-(CH_2)_5$$

$$-NH-COO-CH_3, -NH-COO-CH_2-CH_3, -NH-(CH_2)_3Si(OR)_3$$

$$-S_x-(CH_2)_3Si(OR)_3$$

Organosilanes of the type $(R")_X(RO)_YSi(CH_2)_m-R'$

$$R'' = alkyl$$

$$x+y = 2$$

$$x = 1,2$$

$$y = 1,2$$

 $R' = alkyl$, $aryl (e.g. -C_{6}H_{5})$
 $-C_{4}F_{9}$, $-OCF_{2}-CHF-CF_{3}$, $-C_{6}F_{13}$, $-O-CF_{2}-CHF_{2}$
 $-NH_{2}$, $-N_{3}$, $-SCN$, $-CH=CH_{2}$,
 $-OCC(CH_{3})C = CH_{2}$
 $-OCH_{2}-CH(O)CH_{2}$
 $-NH-CO-N-CO-(CH_{2})_{5}$
 $-NH-COO-CH_{3}$, $-NH-COO-CH_{2}-CH_{3}$, $-NH-(CH_{2})_{3}Si(OR)_{3}$
 $-S_{x}-(CH_{2})_{3}Si(OR)_{3}$

The silane Si 108 [(CH₃O)₃-Si-C₈H₁₇] trimethoxyoctylsilane can preferably be employed as the silanizing agent.

The following substances can be employed in particular as silazanes:

Silazanes of the type:

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and, for example, hexamethyldisilazane.

The following substances can be employed in particular as siloxanes:

Cyclic polysiloxanes of the type D 3, D 4, D 5, e.g. octamethylcyclotetrasiloxane = D 4

Polysiloxanes or silicone oils of the type:

$$Y = 0 = \begin{bmatrix} R \\ \vdots \\ S \\ \vdots \\ R \end{bmatrix} = \begin{bmatrix} R \\ \vdots \\ S \\ \vdots \\ R \end{bmatrix} = \begin{bmatrix} R \\ \vdots \\ R \end{bmatrix} = \begin{bmatrix} R$$

R = alkyl, aryl, $(CH_2)_n - NH_2$, H

R' = alkyl, aryl, $(CH_2)_n - NH_2$, H

S'' = alkyl, aryl, $(CH_2)_n - NH_2$, H

R''' = alkyl, aryl, $(CH_2)_n - NH_2$, H

Y = CH_3 , H, C_nH_{2n+1} where n=1-20

Y = $Si(CH_3)_3$, $Si(CH_3)_2H$ $Si(CH_3)_2OH$, $Si(CH_3)_2(OCH_3)$ Si(CH_3) 2 (C_nH_{2n+1}) where n=1-20

m = 0,1,2,3,...

n = 0,1,2,3,...

u = 0,1,2,3,...

- The silanization can be carried out by a procedure in which the granules are sprayed with the silanizing agent, which can optionally be dissolved in an organic solvent, such as, for example, ethanol, and the mixture is then heat-treated at a temperature of 105 to 400°C over a period of 1 to 6 h.
- An alternative method of the silanization of the granules can be carried out by a procedure in which the granules are treated with the silanizing agent in vapour form and the mixture is then heat-treated at a temperature of 200 to 800°C over a period of 0.5 to 6 h. The heat treatment can be carried out under an inert gas, such as, for example, nitrogen.

The silanization can be carried out continuously or batchwise in heatable mixers and dryers with spray devices. Suitable devices can be, for example: plough share mixers or plate, fluidized bed or flow-bed dryers.

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By varying the starting substances, the conditions during spraying, the heat treatment and the silanization, the physico-chemical parameters of the granules, such as the specific surface area, the particle size distribution, the pore volume, the tamped density and the silanol group concentration, pore distribution and pH, can be modified within the stated limits.

The invention also provides:

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- Dyestuff comprising granules based on pyrogenically prepared silicon dioxide.
 - Antioxidant comprising granules based on pyrogenically prepared silicon dioxide.
 - Preservative comprising granules based on pyrogenically prepared silicon dioxide.
- 15 Emulsifier comprising granules based on pyrogenically prepared silicon dioxide.
 - Gelling agent comprising granules based on pyrogenically prepared silicon dioxide.
- Thickener comprising granules based on pyrogenically prepared silicon dioxide.
 - Binder comprising granules based on pyrogenically prepared silicon dioxide.
 - Stabilizer comprising granules based on pyrogenically prepared silicon dioxide.
- 25 Alkali comprising granules based on pyrogenically prepared silicon dioxide.
 - Acids comprising granules based on pyrogenically prepared silicon dioxide.

- Salts comprising granules based on pyrogenically prepared silicon dioxide.
- Antilumping agent comprising granules based on pyrogenically prepared silicon dioxide.
- 5 Flavour intensifier comprising granules based on pyrogenically prepared silicon dioxide.
 - Sweetener comprising granules based on pyrogenically prepared silicon dioxide.
- Aroma comprising granules based on pyrogenically prepared silicon dioxide.
 - Feedstuffs additives comprising granules based on pyrogenically prepared silicon dioxide.
 - Chemical intermediates comprising granules based on pyrogenically prepared silicon dioxide.
- 15 Plant protection agents comprising granules based on pyrogenically prepared silicon dioxide.
 - Herbicides comprising granules based on pyrogenically prepared silicon dioxide.
- Insecticides comprising granules based on pyrogenically prepared silicon dioxide.
 - Fungicides comprising granules based on pyrogenically prepared silicon dioxide.

Foodstuffs additives can be:

Dyestuffs, such as, for example:

	E100 Curcumin
	E101 Riboflavin, Lactoflavin
5	E102 Tartrazine
	E104 Quinoline Yellow
	E110 Sunset Yellow S (azo dyestuff)
	E120 Carminic acid, Cochineal
	E122 Azorubine (azo dyestuff)
10	E123 Amaranth (azo dyestuff)
	E124 Ponceau 4R (azo dyestuff)
•	E127 Erythrosine
	E131 Patent Blue V
	E132 Indigotine, Indigo Carmine
15	E140 Chlorophylls a + b
	E141 Chlorophylls und Chlorophyllins,
	copper complexes
•	E142 Acid Brilliant Green BS
•	E150 Caramel, Sugar colour, Rum colour
20	E151 Brilliant Black BN (azo dyestuff)
	E153 Charcoal, medicinal
	E160 Carotenoids
	E160a Beta-Carotene, Gamma-Carotene
	E160b Bixin, Norbixin, (Annatto), Orlean
25	E160c Capsanthin, Capsorubin
	E160d Lycopene
	E160e Beta-Apo-8'-Carotenal (C30)
	E160f Beta-Apo-8'-Carotenoic Acid Ethyl Ester
	E161 Xanthophylls
30	E161a Flavoxanthin
	E161b Lutein
	E161c Cryptoxanthin
	E161d Rubixanthin
	E161e Violaxanthin
35	E161f Rhodoxanthin

E161g Canthaxanthin

E162 Betanin, Beetroot Red

	E163 Anthocyan	
	E172 Iron oxide, iron hydroxide	
5	E173 Aluminium	
	E174 Silver	
	E175 Gold	
	E180 Pigment Rubine BK, Lithol Rubine	
0	Antioxidants can be:	
- 0		
	E220 Sulfurous acid, sulfur dioxide	
	E221 Sodium sulfite	
	E222 Sodium hydrogen sulfite	•
	E223 Sodium disulfite	,
L 5	E224 Potassium disulfite	
	E300 Ascorbic acid	•
	E301 Sodium ascorbate	
	E302 Calcium ascorbate	
	E304 Ascorbyl palmitate	
20	E306 Tocopherol-containing extracts of	natural
	origin	
	E307 alpha-Tocopherol	·
	E308 gamma-Tocopherol	
	E309 delta-Tocopherol	
25	E310 Propyl gallate	
	E311 Octyl gallate	
	E312 Dodecyl gallate	
	E320 Butylhydroxyanisole (BHA)	
	E321 Butylhydroxytoluene (BHT)	
30	E330 Citric acid	
	E331 Sodium citrate	
	E332 Potassium citrate	
	E333 Calcium citrate	
	E472c Citric acid esters	
35	Ethoxiquin	

Preservatives can be:

	E200 Sorbic acid
•	E201 Sodium sorbate
	E202 Potassium sorbate
5	E203 Calcium sorbate
	E210 Benzoic acid
	E211 Sodium benzoate
	E212 Potassium benzoate
•	E213 Calcium benzoate
10 .	E214 Ethyl 4-hydroxybenzoate
•	E215 Ethyl 4-hydroxybenzoate, sodium salt
	E216 Propyl 4-hydroxybenzoate
	E217 Propyl 4-hydroxybenzoate, sodium salt
	E218 Methyl 4-hydroxybenzoate
15	E219 Methyl 4-hydroxybenzoate, sodium salt
•	E220 Sulfurous acid, sulfur dioxide
•	E221 Sodium sulfite
	E222 Sodium hydrogen sulfite
	E223 Sodium disulfite
20	E224 Potassium disulfite
	E236 Formic acid
•	E280 Propionic acid
	E281 Sodium propionate
	E282 Calcium propionate
25	E283 Potassium propionate

Emulsifiers can be:

E322 Lecithin

E442 Ammonium salts of phosphatidic acids

E471 Edible fatty acids, mono- and

diglycerides

E472 Esters of E471

E472a Acetic acid esters

E472b Lactic acid esters

E472c Citric acid esters

E472d Tartaric acid esters

	ullet
	E472e Diacetyltartaric acid esters
	E472f Tartaric-acetic acid esters
	E473 Sucrose esters of edible fatty acids
•	E474 Sugar glycerides
5	E475 Polyglycerol esters of edible fatty acids
	E476 Polyglycerol esters of polycondensed
	ricinoleic acid
	E477 Propylene glycol esters of edible fatty
	acids
10 .	E481 Sodium stearoyllactylate
	E482 Calcium stearoyllactylate
	E487 Sdoium lauryl sulfate
•	

Gelling agents, thickeners and binders and stabilizers can be:

	·
•	E400 Alginic acid
	E401 Sodium alginate
•	E402 Potassium alginate
	E403 Ammonium alginate
20	E404 Calcium alginate
•	E405 Propylene glycol alginate
	E406 Agar-agar
	E407 Carrageenan
•	E410 Carob bean flour
25	E412 Guar flour
	E413 Tragacanth
	E414 Gum arabic
•	E415 Xanthan
	E416 Karaya gum
30	E417 Tara gum
	E440 Pectins
	E460a Cellulose, microcrystalline
•	E460b Cellulose, powdered
	E461 Methylcellulose
35	E463 Hydroxypropylcellulose

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•	E464 Hydroxypropylmethycellulose
	E465 Methylethylcellulose
	E466 Carboxymethylcellulose
	E551 Silicon dioxide
5	E1411 Di-starch phosphate I
	E1412 Di-starch phosphate II
	E1413 Di-starch phosphate, phosphated
-	E1414 Di-starch phosphate, acetylated
	E1420 Mono-starch acetate I
10	E1421 Mono-starch acetate II
	E1422 Di-starch adipate, acetylated
	E1423 Di-starch glycerol, acetylated
•	E1430 Di-starch glycerol
•	E1440 Hydroxypropyl-starch
15	E1441 Hydroxypropyl-di-starch glycerol
	E1442 Hydroxypropyl-di-starch phosphate

Alkalis, acids and salts can be:

	E170	Calcium carbonate
20 .	E260	Acetic acid
	E261	Potassium acetate
·	E262	Sodium diacetate
•	E263	Calcium diacetate
	E270	Lactic acid
25	E296	Malic acid
	E325	Sodium lactate
•	E326	Potassium lactate
	E327	Calcium lactate
•	E330	Citric acid
30	E331	Sodium citrate
	E332	Potassium citrate
	E333	Calcium citrate
	E334	Tartaric acid
	E335	Sodium tartrate
35	E336	Potassium tartrate
		•

•	E337 Potassium sodium tartrate
	E354 Calcium tartrate
	E338 Orthophosphoric acid
	E339 Sodium orthophosphate
5	E340 Potassium orthophosphate
•	E341 Calcium orthophosphate
	E343 Magnesium orthophosphate
	E350 Sodium malate
	E351 Potassium malate
10	E352 Calcium malate
	E450 Salts of di-, tri- and polyphosphoric
•	acid (di-, tri- and polyphosphates)
	E500 Sodium carbonate
	E501 Potassium carbonate
15	E503 Ammonium carbonate
	E504 Magnesium carbonate
	E507 Hydrochloric acid
	E508 Potassium chloride
	E509 Calcium chloride
20	E510 Ammonium chloride
	E514 Sodium sulfate
	E515 Potassium sulfate
	E516 Calcium sulfate
	E524 Sodium hydroxide
25	E525 Potassium hydroxide
	E526 Calcium hydroxide
	E527 Ammonium hydroxide
	E528 Magnesium hydroxide
	E529 Calcium oxide
30	E530 Magnesium oxide
	E541 Sodium aluminium phosphate
	E574 Gluconic acid
	E575 Glucono-delta-lactone
	E576 Sodium gluconate
35	E577 Potassium gluconate
	E578 Calcium gluconate

Antilumping agents can be:

E170 Calcium carbonate
E341 Calcium orthophosphate
E470 Edible fatty acids, sodium, potassium and
calcium salts
E504 Magnesium carbonate
E535 Sodium ferrocyanide
E536 Potassium ferrocyanide
E538 Calcium ferrocyanide

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Flavour intensifiers can be:

E621 Sodium glutamate
E622 Potassium glutamate
E623 Calcium glutamate
E624 Magnesium glutamate
E625 Ammonium glutamate
E627 Sodium guanylate
E628 Potassium guanylate
E629 Calcium guanylate
E630 5'-Inosinic acid
E631 Sodium inosinate
E632 Potassium inosinate
E633 Calcium inosinate

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Sweeteners can be:

E950 Acesulfame-K
E951 Aspartame
E952 Cyclamate
Sep54 Saccharin
E957 Thaumatin

Aromas:

Abriceine Acetanisole cryst. Acetophenone pure Agar wood D50092NS · 5 Agrumen aldehyde 6947L Agrumex HC Agrumex LC Agrumovert 10897 C/J Aldehyde C 6 nat. 10 Aldehyde C11 MOA Aldehyde C12 MNA Aldehyde C14 so-called Aldehyde C16 so-called Aldehyde C18 so-called/Abricolin 15 Alcohol C 6 nat. Alcohol C 8 Alcohol C 9 Alcohol C10 Alcohol C12 20 Allinat/Allyl isothiocyanate Allinat/Allyl isothiocyanate (stab.) Allyl caproate Allyl caproate kosher Allyl cyclohexylpropionate 25 Allyl heptylate Allyl phenoxyacetate Amarocit ® Ambre 83LN DB10028 Ambrebois D50407 30 Ambrettia C Ambrettolide Ambrinol S Ambroxid cryst. Ananas Coeur D50214 35 Anethol NPU 21/22°C

	Anethol supra 21.5°C
	Anisaldehyde pure
	Anisyl alcohol
	Anisole
5	Anisyl acetate
•	Apple 74180C PM
	Apriconia 28855P extra PM
	Baldrian Identoil B
	Basilicum Synthessence
10	Bay Identoil
•	Benzalacetone
•	Benzaldehyde
	Benzaldehyde dd
	Benzophenone cryst.
15	Benzyl acetate
•	Benzyl acetone
	Benzyl alcohol dd
	Benzyl alcohol FR
	Benzyl benzoate H&R
20	Benzyl benzoate M
	Benzyl cinnamate
	Benzyl formate
	Benzyl propionate
	Benzyl salicylate
25	Bergamot Identoil colourless
	Bergamot Synthessence Afric.
	Blackberry D50260E
	Bois de Cachemire D50008
	Bois Doux 78008SP PM
30	Boronal
•	Butyric acid nat.
	Butyl butyrate
	Cacao et Chocolat D50546B
	Cajeput Identoil
35	Calmus Synthessence asarone-free
	Cananga Identoil
	Capric acid nat.

	Caproic acid nat.
	Caramel acetate
	Cardamom R Identoil
-	Cardamom Synthessence
5	Cassia Identoil
·	Cassia Identoil B dark
•	Cassis D50060B
	Cedar Leaves Identoil
·	Chloracetophenone para
10	Chrysantheme
	Cinnamyl acetate
	Citral FF
	Citron R
	Citrone Synthessence FF
15	Citronella Identoil
	Citronell Identoil
	Citronellyl tiglinate
•	Citronitrile
. ,	Citrowanil® B
20	Citrozone D50620B
	Citrylal
	Citrylal E
	Clarifruit D50757
	Clarion Base D50774
25	Coriander Identoil
	Corps 98N DB10025
	Corps Racine VS
·	Costus Synthessence
•	Coumarone
30	Cumin Synthessence
	Cypress Identoil
	Damascenone beta nat. 1% in EtOH
	Datilat ·
	Decalactone gamma nat.
35	Decalyl acetate beta
:	Diacetyl nat.
•	Dibenzosuberenone

Dibenzosuberone Dibenzyl ether Diethyl phthalate (DEP) Dihydrocoumarin Dimethyl anthranilate 5 Dimethyl benzyl carbinyl butyrate Dimethyl sulfide nat. Diphenyl oxide Silver Fir Needle Identoil Silver Fir Needle Identoil B 10 Oak Moss Resin D50342 Strawberry D50026C Acetic acid nat. Estragon Identoil Ethoxyfuranone 15 Ethyl 2-methylbutyrate nat. Ethyl 2-methylbutyrate Ethyl acetate nat. Ethyl acetoacetate 20 Ethyl benzoate Ethyl butyrate Ethyl butyrate nat. Ethyl caproate kosher Ethyl caproate nat. 25 Ethyl caprylate Ethyl caprylate nat. Ethyl cinnamate Ethyl formate Ethyl heptylate Ethyl isovalerate 30 Ethyl phenylacetate Ethyl propionate Ethyl salicylat Eucalyptol Eucalyptus Oil Globulus BP 35 Eugenol Eugenol methyl ether

Farenal Fennel oil techn. Feuilles de Tomate 79569PM Spruce Green 8001S 5 Spruce Needle Identoil B sib. Spruce Needle Identoil sib. Filbertone G Fir Balsam DM Fleur de Cassis SBU PM 10 Floropal Florophyll 10183 Fragolane Framboson 10583F Frutinat Galbanum Synthessence 15 Galbanum Synthresin B Geranium Identoil Afric. Geranium Identoil Bourbon Geranyl tiglinate Globalide 100% 20 Globanone 50% DEP Globanone 50% DPG Globanone 50% IPM Grapefruit D50075N 25 Grapefruit Identoil D61286G Green Honey Melon D50315 Guave 10875N Helichrysum Synthessence Herbaflorat 30 Hexyl acetate Hexyl acetate nat. Hexyl salicylate Hydrocitronitrile Indian Spice 10898 35 Indoflor H&R cryst. Indole FF Ginger oil spec. D40393S

Ionone pure 100% Iris Synthresin H&R Irolene P Isoamyl acetate G Isoamyl acetate nat. Isoamyl butyrate Isoamyl butyrate nat. Isoamyl isobutyrate nat. Isoamyl isovalerate 10 Isoananate Isobornyl acetate Isobutyric acid nat. Isobutyl acetat nat. Isobutylquinoline Isobutylquinoline 54 15 Isoeugenol methyl ether Isotabac naturelle LN DB10038 Jasmaprunat Camomile Identoil blue Camomile rom. Synthessence 20 Pine Needle Identoil Pine Needle Identoil B Pine Needle Identoil B P Kiwi D50195PM Cresol methyl ether para 25 Lactojasmon Lavandin Identoil 30/32% Lavandin Identoil type French 30/32% Lavandin Provence D50817 Lavender Identoil type Mt. Blanc 40/42% 30 Lavender oil type Mt. Blanc 40/42% Leguminal Limonene d pure Loganberry D50398N PM 35 Bay Leaf Oil D50286 Mace Oil extra Macrolide®

Macrolide® supra Madranol Magnolan Majantol 5 Mandaril Manderine Synthessence Mango D50436PM Maracuja D50042E PM Marjoliane N DB10018 10 Mayciane N DB10023 Melissa Identoil German so-called Menthol D dist. Menthol liquid Menthol rac. 15 Menthol rac. PH Menthol-1 dest. Menthol-1 H&R compacted Menthol oil Menthone-1/Isomenthone-d 20 Menthyl acetate-1 Metaxa D50247C Methyl ethylpyrazine-2,3 Methyl 2-methylbutyrate Methylacetophenone para 25 Methylacetophenone para supra Methyl anthranilate Methyl benzoate H&R Methyl benzoate techn. pure Methyl betanaphthyl ketone cryst. 30 Methylbutyric acid-2 nat. Methyl cinnamate Methyl phenylacetate Methyl salicylate Methyl cinnamaldehyde alpha 35 Miel Blanc N DB10024 Musk Seed Synthessence Mugetanol

	Mugofleur D50444PM
	Clary Sage Identoil
	Clary Sage Identoil B
	Carnation Flower Identoil
5	Clove Leaf Identoil dark
	Clove Leaf Oil deg.
	Neononyl acetate
	Neroli Identoil
	Nerolin Yara Yara cryst.
10	Neroli oil 4663
	Olibanum Synthresin
	Orange Identoil TSA
	Orange oil spec. D40393P
	Origanum Identoil
15	Oryclon extra
	Oryclon special
	Osmanthia 353
	Ozonil
	Palisandal
20	Palisandin
	Palmarosa Synthessence
	Pastinak Synthessence
	Patchouli Synthessence N
	Patchouli oil deg. DM
25	Pear D50313A PM
	Peru Balsam Identoil
	Peru balsam art. H&R
	Petitgrain Bigarade Synthessence
	Petitgrain Identoil R
30	Peach D40110PM
•	Plum D50424
	Phenirate
	Phenoxyethyl alcohol/arosol
•	Phenylacetaldehyde 100%
35	Phenylacetaldehyde dimethyl acetal
	Phenylethyl acetate
	Phenylethyl alcohol benzyl alcohol-free

	Phenylethyl alcohol pure
	Phenylethyl cinnamate cryst.
•	Phenylethyl isobutyrate
	Phenylethyl phenylacetate
5	Phenylpropyl alcohol
	Pimento Identoil
•	Pineapple acetate
	Poivre Coeur H&R PM
	Poivron N DB10029
10	Prenyl acetate
	Prenyl salicylate
	Profarnesal
	Projasmon P
•	Propionic acid nat
15	Propyl acetate nat.
-	Prunol N DB10027
	Pyroprunat
•	Rain Forest D50339C PM
	Resedafol
20	Rosaphen
	Rose Booster D50221A
	Rose F50048R PG
	Rosemary Identoil
	Rosemary Identoil Spanish
25	Rosewood Braz. Identoil
	Sage Identoil Span.
	Sage Identoil Span.
	Sandalwood S.E.A. D50820
	Sandel 80
30	Sandel extra
•	Sandel Forte
	Sandel H&R
•	Sandel H&R ECO
	Sandel H&R super
35	Sandel SP
•	Sandel type East Ind.
	Sandalwood type East Ind.
	•

	Sandolene H&R
	Spike Identoil
	Styrax Identoil D50186
•	Styrenyl acetate
5	Sweet Amber D50807
	Tobacco aroma H&R D50799
	Teatree D50780A
	Thyme Identoil
,	Thyme red Identoil
10	Thyme Synthabsolue
	Thymol dist.
•	Thymol cryst. H&R
	Thymol cryst. PH
	Tonca Synthresin
15	Vanillin nat.
•	Verbena Identoil type French
	Verdeflora D50375D
	Verdural F
•	Vertocitral
20 .	Vertocitral C
•	Vertosine
•	Vetiver Identoil J
•	Juniper berry Identoil 10900
	Juniper berry Synthessence
25	Willow fragrance 6103CB HG
	Wintergreen oil
	Ylang 10372 MT
	Ylang Ylang Identoil Bourbon I
	Ylang Ylang Identoil Bourbon II
30 .	Ylang Ylang Identoil Bourbon III
•	Cinnamaldehyde
	Cinnamaldehyde nat.
	Cinnamyl alcohol
	Cinnamon leaf Identoil
35	Cinnamon bark Identoil

Feedstuffs additives can be:

Choline chloride solution Vitamin E acetate Formic acid Acetic acid Propionic acid Phosphoric acid Fat concentrates Ethoxiquin Molasses 10 Hop extract Tagetese extract Lecithin Whey Calcium formiate 15 Urea. Milk substitute Trace elements

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Chemical intermediates can be:

Vitamins

1,2-Propylene glycol
Acrylic acid
Adipic acid

25
Adipic anhydride
Formic acid
Formic anhydride
Benzoic acid
Succinic acid
Succinic acid
Butanoic acid
Butanoic anhydride
Caproic acid
Dimer fatty acid
Dimer fatty acid anhydride

	Dipentaerythritol
	Erucic acid
	Acetic acid
	Acetic anhydride
5	Ethylene glycol
	Fumaric acid
	Glutaric acid
	Glycerol
-	Isophthalic acid
. 0	Isophthalic anhydride
	Lauric acid
	Linolenic acid
	Linoleic acid
	Maleic acid
.5	Maleic anhydride
	Malonic acid
	Myristic acid
	Oleic acid
	Oxalic acid
20	Palmitic acid
	Pentaerythritol
	Phthalic acid
	Phthalic anhydride
·	Propionic acid
25	Stearic acid
	Terephthalic acid
•	Terephthalic anhydride
	Trimethylolpropane
	Valeric acid
30	Bisphenol A
	Epichlorohydrin
•	o-Cresol
•	Phenol novolaks
	Styrene
35	α-Methylstyrene
	Vinyltoluene
	Methyl methacrylate

	Divinylbenzene
	Diallyl phthalate
	Diisocyanates
	Toluene-diisocyanates
5	Cyclohexanone
	Methylcyclohexanone
	Acetone
	Butanone
	Acetophenone
10	Indene
•	Coumarone (benzofuran)
•	2-Methylindene
	2-Methylcoumarone
•	Methylstyrene
15	Cyclopentadiene
•	Dicyclopentadiene
•	Heteropolysaccharides
	Arabinose
	Galactose
20	Glucoronic acid
•	Mannose
•	Rhamnose
	Xylose
•	Resinol acids
25	Resinols
	Resinotannols
,	Resenes
	Terpenes
•	Diterpenes
30	Triterpenes
•	Sesquiterpenes
	Resin esters
	Resin soaps
	Alcohols
35	Phenol derivatives
	Hydroquinone derivatives
	Ouinoline derivatives

Naturally occurring resins:

Acaroid resin Asa foetida Benzoin resin Amber Bitumen Canada balsam China lacquer Copaiva balsam Dammar resin 10 Dragon's blood resin Elemi Galbanum Gutti Jalap resin 15 Japan lacquer Kauri copal Colophony Copal Labdanum 20 Manila copal Mastix Myrrh Olibanum 25 Opoponax Pernambuco balsam Peru balsam Sandarac Shellac 30 Styrax Tolu balsam Terpentine

Synthetic resins:

35

Hydrocarbon resins Urea resins

Alkyd resins Epoxy resins Melamine resins Melamine-formaldehyde resins Hexamethylolmelamine resins 5 Melamine-phenol resins Melamine-urea resins Phenolic resins Polyester resins Unsaturated polyester resins 10 Polyurethane resins Ketone resins Coumarone-indene resins Isocyanate resins Polyamide resins 15 Terpene-phenol resins Epoxy resins Rubber

Additives:

Wetting agents 20 Desiccants Antifloating agents Antiskinning agents Hardening accelerators Hardening retardants 25 Expanding agents Sealants Water softeners Deoxygenating agents Buffers 30 Polishing agents Antiageing agents Antioxidants Antiozonants Plasticizers 35

Deodorizers Inhibitors Passivating agents Pickling inhibitors Anticorrosion agents Antistatics Stabilizers Release agents Lubricants Flameproofing agents 10 UV absorbers Antiknocking agents Corrosion inhibitors Metal deactivators Carburettor cleaning agents **15** · Residue converters Antiicing agents Pour point depressors Defoamers Lubricity improvers Optical brighteners

Antifoams:

25

Anionic surfactants
Polyethylene ethers
Polypropylene glycol ethers
Pluronic®
Mixed ethers

Inorg. peroxides:

Hydrogen peroxide
Lithium peroxide
Sodium peroxide
Calcium peroxide
Strontium peroxide
Barium peroxide

Org. peroxides:

Di-tert-butyl peroxide

Dibenzoyl peroxide

Per-acids

Per-acid esters

Ketone peroxides

Epidioxides

Ascaridol

Ergosterol peroxide

Stabilizers:

Ethylenediaminetetraacetic acid Magnesium silicate

20 Plasticizers:

Trimellitic acid
Phosphoric acid esters
Azelaic acid esters

Sebacic acid esters
Chloroparaffins
Dioctyl phthalate
Bis-(2-ethylhexyl) phthalate
Diisononyl phthalate
Diisodocecyl phthalate
Phthalic acid esters
Dibutyl phthalate

	Disobutyi phtharate
	Dicyclohexyl phthalate
	Dimethyl phthalate
	Diethyl phthalate
5	Benzyl butyl phthalate
•	Butyl octyl phthalate
· - · · · · · · · · · · · · · · · · · ·	Butyl deyl phthalate
	Dipentyl phthalate
	Dimethylglycol phthalate
1.0	Dicapryl phthalate
	Trimellitic acid esters
	Tris-(2-ethylhexyl) trimellitate
	Dioctyl adipate
	Bis-(2-ethylhexyl) adipate
15	Diisodecyl adipate
	Dibutyl sebacate
	Dioctyl sebacate
•	Bis-(2-ethylhexyl) sebacate
	Azelaic acid
20	Sebacic acid
	1,3-Butanediol
	1,2-Propanediol
•	1,4-Butanediol
	1,6-Hexanediol
25	Tricresyl phosphate
•	Triphenyl phosphate
	Diphenyl cresyl phosphate
	Diphenyl octyl phosphate
	Bis-(2-ethylhexyl)diphenyl phosphate
30	Tris-(2-ethylhexyl) phosphate
	Tris-(2-butoxyethyl) phosphate
•	Butyl oleate
	Butyl stearate
	Triethylene glycol bis-(2-ethylbutyrate)
35	Citric acid esters
	Acetyltributyl citrate

Tartaric acid esters
Lactic acid esters
Epoxystearic acid esters
Epoxidized soya oils
Linseed oils
Benzenesulfonamides
p-Toluenesulfonamides

Free radical interceptors:

Nitrogen monoxide

Bis(trifluoromethyl) nitroxide

Nitroxyl radicals

2,2-Diphenyl-1-picrylhydrazyl

Nitrosobenzene

2-Methyl-2-nitroso-propane

Benzaldehyde tert-butyl nitrone

Wetting agents can be:

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Dimethyloctylphosphine oxide Dimethylnonylphosphine oxide Dimethyldecylphosphine oxide Dimethylundecylphosphine oxide Dimethyldodecylphoshine oxide N,N,-bis(3-D-gluconamidopropyl)cholamide N, N-Bis (3-D-gluconamidopropyl) deoxycholamide Dodecylpoly(oxyethylene glycol ether)s, PEG (23) dodecyl ether, PEG (10) cetyl alcohol PEG (20) cetyl alcohol PEG (10) stearyl alcohol PEG (10) oleyl alcohol PEG (29) oleyl alcohol Polyethylene glycol (10) lauryl ether Polyethylene glycol (8) dodecyl ether

Polyethylene glycol (10) isotridecyl ether

Polyethyleneglycol (15) isotridecylether Ethylphenol-poly(ethylene glycol ether)s Lubrol Thesit Thesit 5 Cetylpyridinium chloride Cetyltrimethylammonium bromide 3-[(3-Cholamidopropyl)dimethylammonio]-1-propanesulfonic acid 3-[(3-Cholamidopropyl)dimethylammonio]-1-10 hydroxypropanesulfonic acid Chenodeoxycholic acid Cholate, Na+ Deoxycholate, Na+ Glycocholate, Na+ 15 Glycodeoxycholate, Na+ Taurocholate, Na+ Taurodehydrocholate, Na+ Taurodeoxycholate, Na+ $\texttt{Cyclohexyl-} n-\texttt{ethyl-}\beta-\texttt{D-maltoside}$ 20 $Cyclohexyl-n-hexyl-\beta-D-maltoside$ Cyclohexyl-n-methyl-\beta-D-maltoside $n-Decyl-\beta-D-maltopyranoside$ n-Dodecyl-beta-D-maltopyranoside $n-Octyl-\beta-D-maltopyranoside$ 25 $n-Undecyl-\beta-D-maltoside$ N, N-Dimethyldecylamine oxide Genaminox KC N, N-Dimethyldodecylamine oxide N-Dodecyl-N, N-(dimethylammonio)butyrate 30 N-Dodecyl-N, N-(dimethylammonio)undecanoate n-Dodecyl-N, N-dimethylglycine N-Octyl-N, N-dimethylammonio-3-propanesulfonate N-Decyl-N, N-dimethylammonio-3-propanesulfonate N-Dodecyl-N, N-dimethylammonio-3-propanesulfonate 35 N-Tetradecyl-N,N-dimethylammonio-3-propanesulfonate Decanoylsucrose

n-Dodecanoylsucrose Octanoylsucrose n-Decyl-β-D-glucopyranoside Dodecyl-β-D-glucopyranoside $n-Heptyl-\beta-D-glucopyranoside$ n-Hexyl-β-D-glucopyranoside $n-Nonyl-\beta-D-glucopyranoside$ $n-Octanoyl-\beta-D-glucosylamine$ n-Octyl-beta-D-glucopyranoside n-Decyl-β-D-thiomaltoside 10 n-Nonyl-beta-D-thiomaltopyranoside N, N-Bis (3-D-gluconamidopropyl) deoxycholamide N, N, -bis (3-D-gluconamidopropyl) cholamide Digitonin Bis(2-ethylhexyl)sodium sulfosuccinate 15 n-Dodecyl-N, N-dimethylglycine 6-0-(N-heptyl-carbamoyl)methyl-α-D-glucopyranoside N-Dodecanoyl-N-methylglycine Lauryl-sulfate Li+ Lauryl-sulfate, Na+ 20 {3-([4-tert-Octyl]-1-propanesulfonic acid, Na+ n-Octanoyl-N-methylglucamide n-Nonanoyl-N-methylglucamide n-Decanoyl-N-methylglucamide Ethylphenol-poly(ethylene glycol ether)s 25 n-Octyl-2-hydroxyethylsulfoxide n-Octyl-2-hydroxyethyl sulfide n-Octyl-rac-2,3-dihydroxypropylsulfone n-octyl-rac-2,3-dihydroxypropylsulfoxide Polyethylene glycol-polypropylene glycol copolymer 30 Pluronic F-127 β-D-Fructopyranosyl-alpha-D-glucopyranoside monodecanoate β-D-Fructopyranosyl-alpha-D-glucopyranoside monododecanoate PEG (9-10) nonylphenol PEG (4.5) p-t-octylphenol 35 PEG (9-10) p-t-octylphenol PEG (9-10) p-t-octylcyclohexyl

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PEG (7-8) p-t-octylphenol PEG (7-8) t-octylcyclohexyl

Plant protection agents can be:

	······································	1		
Herbicides	Insecticides	Fungicides	Other	
2,4-D	Abamectin	Abamectin Acibenzolar		
2,4-DB	Acephate	Azoxystrobin	Chloropicrin	
Acetochlor	Acequinocyl	Benalaxyl	Choline Chloride	
Acifluorfen	Acetamiprid	Benomy1	Cyclanilide	
Aclonifen	Acrinathrin	Bitertanol	Dazomet	
Alachlor	Alanycarb	Bromuconazole	Dichlopropene	
Alloxidim	Aldicarb	Bupirimate	Dikegulac	
Ametryn	Alpha- cypermethrin	Captan	Dimethipin	
Amidosulfuron	Amitraz	Carbendazim	Ethepon	
Aminotriazole	Azinphos-methyl	Carboxin	Flumetralin	
Anilofos	Azocyclotin	Carpropamid	Gibberellic acid	
Asulam	Bacillus thuringiensis	Chlorothalonil	Inabenfide	
Atrazine	Bendiocarb	Chlozolinate	Maleic hydrazide	
Azimsulfuron	Benfuracarb	Copper fungicides	Mepiquat	
Benazolin	Bensultap	Cymoxanil	Metam	
Benfluralin	Benzoximate .	Cyproconazole	Methyl bromide	
Benfuresate	Bifenazate	Cyprodinil	Methyl isothiocyanate	
Bensulforon	Bifentrin	Dichlofluanid	Paclobutrazol	
Bentazone	BPMC (Fenobucarb)	Diclomezine	Prohexadione	
Benzofenap	Bromopropylate	Diethofencarb	Thidiazuron	
Bifenox	Buprofezin	Difenoconazole	Triapenthenol	
Bilanafos .	Cadusafos	Dimethirimol	Tributyl phosphorotri- thioate	
Bispyribac- sodium	Carbaryl	Dimethomorp	Trinexapac-ethyl	
Bromacil	Carbofuran	Diniconazole	Uniconazole	
Bromobuthide	Carbosulfan	Dinocap Fluthiacet 9201 / CGA 248757		
Bromofenoxim	Cartap	Dithianon		

Chinomathionat	Dodemorph	_
Chlorethoxyfos	Dodine	
Chlorfenapyr	Edifenphos	
Chlorfenvinphos	Epoxiconazole	
Chlorfluazuron	Ethaboxam	
Chlormephos	Ethirimol	
Chloropirifos	Etridiazole	
Clofentezine	Famoxadone	
Cycloprothirin	Fenarimol	
Cyfluthrin	Fenbuconazole	
Cyhexatin	Fenhexamid	
Cypermethrin	Fenitropan	
Cyromazine	Fenpiclonil	
Deltamethrin	Fenpropidin	
Demeton-s-methyl	Fenpropimorph	·
Diafenthiuron	Fentin	
Diazinon	Ferimzone	
Dichlorvos	Fluazinam	
Dicofol	Fludioxonil	
Dicrotophos	Fluoroimide	
Diflubenzuron	Fluquinconazole	,
Dimethoate	Flusilazole	
Insecticides	Fungicides	PGR
Disolfoton	Flusulfamide	Aminoethoxy- vinylglycine
Emamectin benzoate	Flutolanil	Prohydrojasmon - PDJ
Endosulfan	Flutriafol	
Esfenvalerate	Folpet	
Ethiofencarb	Fosetyl	
Ethion	Fuberidazole	
Ethoprophos	Furalaxyl	
Etofenprox	Furametpyr	
Etoxazole	Guazatine	
Degraden		
	Chlorethoxyfos Chlorfenapyr Chlorfenvinphos Chlorfluazuron Chlormephos Chloropirifos Chloropirifos Clofentezine Cycloprothirin Cyfluthrin Cyhexatin Cypermethrin Cyromazine Deltamethrin Demeton-s-methyl Diafenthiuron Diazinon Dichlorvos Dicofol Dicrotophos Diflubenzuron Dimethoate Insecticides Disolfoton Emamectin benzoate Endosulfan Esfenvalerate Ethiofencarb Ethion Ethoprophos Etofenprox	Chlorethoxyfos Dodine Chlorfenapyr Edifenphos Chlorfenvinphos Epoxiconazole Chlorfluazuron Ethaboxam Chlormephos Ethirimol Chloropirifos Etridiazole Clofentezine Famoxadone Cycloprothirin Fenarimol Cyfluthrin Fenbuconazole Cypermethrin Fenitropan Cyromazine Fenpiclonil Deltamethrin Fenpropidin Demeton-s-methyl Fenpropimorph Diafenthiuron Fertin Diazinon Ferimzone Dichlorvos Fluazinam Dicofol Fludioxonil Dicrotophos Fluoroimide Diflubenzuron Fluquinconazole Tinsecticides Fungicides Disolfoton Flusulfamide Emamectin benzoate Emdosulfan Flutriafol Esfenvalerate Folpet Ethiofencarb Fosetyl Ethion Fuberidazole Ethoprophos Furalaxyl Etofenprox Furametpyr

Desmetryn	Fenamiphos	Hymexazol	
Dicamba	Fenazaquin	Imazali1	
Dichlobenil	Fenbutatin oxide	Imibenconazole	
Dichlorprp	Fenitrothion	Iminoctadine	
Diclofop	Fenothiocarb	Ipconazole	
Diclosulam	Fenoxycarb	Iprobenfos	·
Difenzoquat	Fenpropthrin	Iprodione	
Diflufenican .	Fenpyroximate	Iprovalicarb	
Diflufenzopyr	Fenthion	Isoprothiolane	
Dimefuron	Fenvalerate	Kasugamŷćin 🗀 📑	
Dimepiperate	Fipronil	Kresoxim-methyl	
Dimethachlor	Flubroythirinate	Mancozeb	
Dimethenamid	Flucycloxuron	Maneb	
Diphenamid	Flucythrinate	Mepanipyrim	
Diquat	Flufenoxuron	Mepronil	
Dithiopyr	Flutenzine	Metalaxyl	
Diuron	Fluvalinate		ali de la companya di managana
Endothal	Formetanate		
EPTC	Formothion	Metiram	
Esprocarb	Fosthiazate	Myclobutanil	
Ethalfluralin	Furathiocarb	Nitrothal-	
:		isopropyl	
Ethametsulfuron		the state of the s	e de la companya de l
	Halofenozide		
Ethoxyfen	•	Oxine-copper	
Ethoxysulfuron	Hexythiazox	Oxolinic acid	
Etobenzanid	Imidacloprid	Oxycarboxin	
Fenoxaprop	Indoxacarb	Pefurazoate	* ,014 - 2.
Flamprop-M	Isofenphos	Penconazole	
Flazasulfuron	Isoprocarb	Pencycuron	·
Fluazifop	Isoxathion	Phthalide	, , , , , , , , , , , , , , , , , , , ,
Flufenacet	Lambda- cyhalothrin	Probenazole	
Flumetsulam	Lindane (Gamma- HCH)	Prochloraz	
Flumiclorac-	Lufenuron	Procymidone	

pentyl			
Flumioxazin	Malathion	Propamocarb	
Fluometuron	Metaldehyde	Propiconazole	·
Fluoroglycofen	Methamidophos	Propineb	
Flupoxam	Methidathion	Pyrazophos	
Flupyrsulfuron	Methiocarb	Pyrifenox	
Herbicides	Insecticides	Fungicides	Nematicides
Flurenol	Methomyl	Pyrimethanil_	ZA3274
Fluridone	Methoprene	Pyroquilon	
Flurochloridone	Methoxyfenozide	Quinoxyfen	
Fluroxypyr	Mevinphos	Quintozene	
Flurtamone	Milbemectin	Spiroxamine	
Fomesafen	Monocrotophos	Streptomycin	·
Glufosinate	Nitenpyram	Sulfur	
Glyphosate	Novaluron	Tebuconazole	
Halosulforon	Omethoate	Tecloftalam	
Haloxyfop	Oxamyl	Tetraconazole	
Imazamethabenz	Oxydemeton-methyl	Thiabendazole	
Imazamox	Parathion	Thilfluzamide	
Imazapic	Parathion-methyl	Thiophanate methyl	
Imazapyr	Permethrin	Thiram	
Imazaquin	Phenthoate	Tolclofos- methyl	
Imazethapyr	Phorate	Tolylfluanid	
Imazosulfuron	Phosalone	Triadimefon	<u> </u>
Isoprothuron	Phosmet	Triadimenol	
Isoxaben	Phosphamidon	Tricyclazole	
Isoxaflutole	Phoxim	Tridemorph	
Lactofen	Pirimicarb	Triflumizole	
Lenacil	Pirimiphos-ethyl	Triforine	
Linuron	Pirimiphos-methyl	Triticonazole	·
MCPA	Profenofos	Validamycin	
MCPA-thioethyl	Propaphos	Vinclozolin	
MCPB	Propargite	Zineb	
Mecoprop	Propoxur	Ziram	

Mefenacet		Cyamidazosulf- amid - IKF-916	. Spr
Metamitron	Pymetrozine		<u>.</u>
Metazachlor	Pyraclofos	Cyamidazosulf- amid- IKF-916	
Methabenzthiazu ron	Pyridaben	-	
Methyl-arsonic acid	Pyridafenthion	Diclocymet - S2900	
Metobromuron	Pyrimidifen	Fenamidone - RPA 407213	
Metolachlor	Pyriproxyfen		
Metosulam	Quinakphos	Fenoxanil - AC382042 /NNF9425	
Metoxuron	Silafluofen		
Metribuzin	Spinosad	Iprovalicarb- SZX722	
Metsulforon	Sulprofos	MA 565	
Molinate	Tebufenozide	Metominostrobin - SSF-126	
Naproanilide	Tebufenpyrad		
Napropamide	Tebupirimfos	MTF-753	
Naptalam	Teflubenzuron	NF-149	
Nicosulfuron	Tefluthrin	NNF-9850	
Norflurazon	Terbufos	Oxpoconazole fumarate - UBF- 910	
Orbencarb	Thiamethoxam		
Oryzalin	Thiocyclam	Picoxystrobin - ZA1963	
Oxadiargyl	Thiodicarb		
Oxadiazon	Thiometon		
Herbicides	Insecticides	Fungicides	
Oxasulfuron	Tralomethrin	Silthiopharm - MON-65500	
Oxyfluorfen	Triazamate		<u>'</u>
Paraquat	Triazophos	Simeconazole - F155	
Pendimethalin	Trichlorfon	Trifloxystrobin	

· · _ · _ · _ · _ · _ · _ · _ · _ ·		- OGA279202
Pentoxazone	Triflumuron	
Phenmedipham	Vamidothion	Zoxamide - RH7281
Picloram	Xylyl methylcarbamate	·
Pretilachlor	Zeta-Cypermethrin	
Primisulfuron	Acetoprole- RPA115782	
Prometryn	AKD 1022	
Propachlor	Chromafenozide- ANS-118	
Propanil		
Propaquizafop	Clothianidin - TI-435	
Propazine	Dinitefuran - MTI-446	
Propyzamide	Ethiprole-RPA 107382	
Prosulfocarb	Fluacrypyrim - NA-83	
Prosulfuron	Flupyrazofos	
Pyraflufen- ethyl	Phosphocarb - BAS301	
Pyrazolinate		
Pyrazosulfuron	Protrifenbute - FMC 111869	
Pyrazoxyfen		
Pyribenzoxim	Thiacloprid - BAYYRC2894	
Pyributicarb		
Pyridate	Tolfenpyrad - OMI-88	·
Pyriminobac- methyl		
Pyrithiobac		
Quinclorac	·	
Quinmerac		
Quinoclamine		·
Quizalofop		

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·	
Quizalofop-P- tefuryl	
Rimsulfuron	
Sethoxydim	
Simazine	
Sulcotrione	
Sulfentrazone	
Sulfometuron	
Sulfosate	· ·
Sulfosulfuron	
Tebuthiuron	
Terbacil	
Terbumeton	
Terbuthylazine	
Terbutryn	
Thenylchlor	-\ .
Thiazopyr	
Thifensulfuron	:"\
Thiobencarb	
Herbicides	
Tralkoxydim	
Triallate	4.
Triasulfuron	.]
Tribenuron	
Triclopyr	
Trifluralin	
Triflusulfuron	
Amicarbazone-BAYMKH3586	
Azafenidin-DPX-R6447	1
Beflubutamid-UBH-820	
Benzfendizone - FMC 143686	
Benzobicyclon -SB-500	
Butafenacil - CGA 276854	
Fentrazamide - BAYYRC2388	
Florasulam - DE570	

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Fluazolate - JV485
Flucarbazone - BAYMKH6562
Flufebpyr-ethyl - S-3453
Foramsulfuron - AEF 130360
Indanofan - MK-243
Iodosulfuron - AEF 115008
Isoxadifen -AEF122006
KPP421
Mesosulfuron - AEF 130060
Mesotrione - ZA1296
MTB-951
OK-9701
Oxaziclomefone-MY-00
Penoxsulam - DE638
Pethoxamid - TKC-94
Picolinofen - AC900001
Propoxycarbazone (proposed) BAYMKH6561
Pyriftalid - CGA279233
Tepraloxydim - BAS620H/NP61EC
Triaziflam - IDH 1105
Trifloxysulfuron (Na salt) - CGA362622
Tritosulfuron

Preferably, however, the silicon dioxide granules employed according to the invention function as a carrier. The present invention therefore also relates to an adsorbate of the silicon dioxide granules described above and at least one of these substances.

The term "adsorbate" as used herein includes the adsorption of a substance not only on to the surface of the silicon dioxide, but also into the pores, as well as the

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"intercalation" into the intergrain volumes. "Adsorbate" can also mean that silicon dioxide granules or fragments thereof envelop solid particles or liquid droplets of the substance. In the latter case the forces of attraction between the particles or droplets are reduced and, for example, the flow properties are improved or the merging of droplets is impeded.

The ratio of amounts of substance to silicon dioxide granules in the adsorbate can be chosen as desired as a function of the properties of the substance and the requirements for the end product. Preferably, however, 0.001 to 200 g of substance are employed per 100 g of silicon dioxide granules, particularly preferably 10 to 150 g.

In a preferred embodiment, granules based on pyrogenically prepared silicon dioxide of average particle diameter from 10 to 120 μm and BET surface area from 40 to 400 m^2/g (determination in accordance with DIN 66 131 with nitrogen) can be used as the silicon dioxide granules.

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The silicon dioxide granules furthermore preferably have the following physico-chemical characteristic data, which are determined as described in EP PS 0 725 037:

Pore volume:

0.5 to 2.5 ml/g

Pore size volume: less than 5% of the total pore volume has a pore diameter of less than 5 nm, remainder meso- and macropores

pH:

3.6 to 8.5

Tamped density:

220 to 700 g/l.

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Granules which are suitable for the use according to the invention and the preparation thereof are described, for example, in EP OS 0 727 037.

An example of a process for the preparation of the adsorbate according to the invention comprises:

Melting of the substance(s) to be adsorbed, chosen from foodstuffs additives, such as dyestuffs, antioxidants, preservatives, emulsifiers, gelling agents, thickeners and binders, stabilizers, alkalis, acids, salts, antilumping agents, flavour intensifiers, sweeteners, aromas, feedstuffs additives, chemical intermediates and plant protection agents, such as, for example, herbicides, insecticides and fungicides, or distribution, i.e. dissolving, suspending or emulsifying, thereof in a solvent;

mixing of the granules based on pyrogenically prepared silicon dioxide with the mixture from step (a); and where appropriate removal of the solvent.

"Solvent" also includes mixtures of several different solvents. It goes without saying, furthermore, that 20 substances which are already liquid at room temperature can be subjected to the mixing in step (b) without prior processing, since in this case the "melting operation" has already taken place. Mixing step (b) can be carried out either by adding the mixture from step (a) to the silicon 25 dioxide granules, for example by spraying on, or vice versa. In both cases, the addition can be made in one amount or in portions. The duration of the mixing in step (b) depends here above all on the adsorption properties of the substance to be adsorbed on the silica surface. If a 30 solvent is present, step (a) and (b) are carried out at a temperature which lies between the freezing and boiling point of the solvent. The solvent, where appropriate in

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excess, is preferably removed in step (c) at elevated temperature and/or under reduced pressure.

The removal of the solvent in step (c) can also be carried out by spray drying or fluidized bed drying, shaping taking place at the same time. In the case of a granule-containing melt, the shaping process can accordingly be an extrusion.

The adsorbates according to the invention can be used for the preparation of powders, liquids, foams, sprays, gels, creams, ointments, pastes, sticks and tablets.

The adsorbates according to the invention can additionally be shaped. They can be processed, for example, to pellets, larger granules, extrudates etc.

The advantage of the adsorbates according to the invention lies in their excellent flow properties, the low water content and the high purity of the starting granules. They offer a very good possibility for dispersing substances which are difficult to meter, and are easy to handle.

When handling the adsorbates, the hazard potential to the administering person during use on toxic substances, such as plant protection agents or aggressive skin-irritating substances, can be reduced significantly.

When the adsorbate according to the invention is used, a uniform distribution of the active compound can be achieved.

The invention is now to be explained in more detail with the aid of examples.

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Preparation of granules based on pyrogenically prepared silicon dioxide

The pyrogenically prepared silicon dioxide AEROSIL 300, commercially obtainable from Degussa AG, is used as the starting compound.

The pyrogenically prepared silicon dioxide is dispersed in completely demineralized water. A dispersing unit which operates by the rotor/stator principle is used here. The suspension formed is spray dried. The finished product is separated off via a filter or cyclone. The heat treatment of the spray granules is carried out in a muffle oven.

The preparation parameters are shown in table 1.

Table 1

Starting SiO ₂	·	AEROSIL 300
Spray drying data		
Amount of H ₂ O	(kg)	100
Amount of SiO ₂	(kg)	10
Atomization with	•	disc atomizer
Operating temperature	(ºC)	480°C
Waste air temperature	(°C)	103°C
Separation		filter
Physico-chemical data		
BET surface area (m^2/g)	298
Particle size d ⁵⁰ (μ m)	3.0
Tamped volume (g/l)	283
рН		4.7

Examples

1. Starting materials

1.1 Model liquids

Vitamin E acetate, silicone oil, paraffin oil and eucalyptus oil are used as model liquids for the fields of use according to the invention. Vitamin E acetate is used, for example, in the nutrition of animals and humans, and eucalyptus oil as an aromatic or aroma substance.

Example	Product	Product name	Manufacturer
1.	Vitamin E acetate		BASF
2	Silicone oil	Silicon Fluid 345	Dow Corning
3	Paraffin oil	Paraffinöl dünnflüssig	Merck
4	Eucalyptus	Oleum Eucalypti 80-85%	Caelo

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1.2 Carrier silicas

Silica	Loss on drying (wt.%)	Loss on ignition (wt.%)	SiO ₂ content (wt.%)	Slope angle (°)	Bulk density (g/l)
Example 1-4 AEROPERL® 300/30 (Degussa)	1.7	2.1	99.9	34.97	232.8
Comparison example 1 SIPERNAT® 22 (Degussa)	4. 8	4.4	98.0	38.99	211
Comparison example 2 SIPERNAT® 50 (Degussa)	4.5	4.9	98.5	52.67	136.67
Comparison example 3 Syloid 244 FP (Grace)	5.9	3.9	nđ	50	92

Granulated pyrogenic silica (AEROPERL® 300/30) has a significantly lower water content (loss on drying and ignition) and a higher silicon dioxide content than the silicas used in the comparison examples. Furthermore, it is free from sulfates, typical impurities of precipitated silica and silica gels, and has the best flowability (the lowest slope angle).

10 2. Procedure:

50 g of carrier silica are initially introduced into a 2 litre three-necked flask equipped with a blade stirrer.
50 g of the model liquids from examples 1-4 are added dropwise from a dropping funnel in the course of 60 minutes, while stirring at a stirrer speed of 100 revolutions / minute. Comparison examples 1-3 are carried out with eucalyptus oil. The liquid-silica adsorbates are

then sieved manually three times through a 0.8 mm sieve and left to stand overnight in a closed screw-cap glass bottle. The following day, the liquid-silica adsorbates are characterized by the following methods:

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Flow rating by means of glass flow vessels in accordance with the publication series Pigmente No. 31 "AEROSIL zur Verbesserung des Fließverhaltens pulverförmiger Substanzen", Degussa AG, Düsseldorf.

- Poured cone height (cm) or slope angle (°) in accordance with the publication series Pigmente No. 31. The slope angle is obtained from the poured cone height via the equation
 - tan(slope angle) = (poured cone height/0.5 cone diameter)

15 Bulk density (g/l) in accordance with DIN standard 6613.

3. Results

•	Flow rating	Slope angle (°)	Bulk density (g/l)
Example 1	2	30.1	501
AEROPERL / Vitam. E			
Example 2	2	37.2	475
AEROPERL / Silicone oil	·	·	
Example 3	2	38.7	497
AEROPERL / Paraffin oil			
Example 4	2	37.2	594
AEROPERL / Eucalyptus oil			·
Comparison example 1	. 3	46.0	450
SIPERNAT 22	· · · .	·	
Comparison example 2	4	63.9	353
SIPERNAT 50			
Comparison example 3	· 6 .	56.7	201
Syloid FP 244			

The liquid-silica adsorbates prepared with granulated pyrogenic silica (AEROPERL® 300/30) are distinguished by a good flowability (flow rating 2, slope angle < 40°C). In contrast, the liquid-silica adsorbates from comparison examples 1 to 3 show a significantly lower flowability. The latter moreover have significantly lower bulk densities.

Liquid-silica adsorbates with a good flowability and high bulk volume are advantageous for carrier uses. Furthermore, carrier silicas should have the lowest possible water content and should be very pure, in order to avoid decomposition of the adsorbed liquids under the (catalytic) influence of water or impurities, such as, for example,

sulfates. The experiments show that granulated pyrogenic silica meets all these requirements.

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Patent claims:

- Use of granules based on pyrogenically prepared silicon dioxide as a carrier for substances chosen from the group consisting of foodstuffs additives, such as dyestuffs, antioxidants, preservatives, emulsifiers, gelling agents, thickeners and binders, stabilizers, alkalis, acids, salts, antilumping agents, flavour intensifiers, sweeteners, aromas, feedstuffs additives, chemical intermediates and plant protection agents, such as, for example, herbicides, insecticides and fungicides.
 - 2. Use according to claim 1, characterized in that the silicon dioxide granules are silanized.
- 3. Adsorbate of granules based on pyrogenically prepared silicon dioxide and at least one substance chosen from the group consisting of foodstuffs additives, such as dyestuffs, antioxidants, preservatives, emulsifiers, gelling agents, thickeners and binders, stabilizers, alkalis, acids, salts, antilumping agents, flavour intensifiers, sweeteners, aromas, feedstuffs additives, chemical intermediates and plant protection agents, such as herbicides, insecticides and fungicides.
 - 4. Adsorbate according to claim 3, characterized in that the silicon dioxide granules are silanized.
- 5. Dyestuff comprising granules based on pyrogenically prepared silicon dioxide.
 - 6. Antioxidant comprising granules based on pyrogenically prepared silicon dioxide.
- 7. Preservative comprising granules based on pyrogenically prepared silicon dioxide.

- 8. Emulsifier comprising granules based on pyrogenically prepared silicon dioxide.
- 9. Gelling agent comprising granules based on pyrogenically prepared silicon dioxide.
- 5 10. Thickener comprising granules based on pyrogenically prepared silicon dioxide.
 - 11. Binder comprising granules based on pyrogenically prepared silicon dioxide.
- 12. Stabilizer comprising granules based on pyrogenically prepared silicon dioxide.
 - 13. Alkali comprising granules based on pyrogenically prepared silicon dioxide.
 - 14. Acids comprising granules based on pyrogenically prepared silicon dioxide.
- 15 15. Salts comprising granules based on pyrogenically prepared silicon dioxide.
 - 16. Antilumping agent comprising granules based on pyrogenically prepared silicon dioxide.
- 17. Flavour intensifier comprising granules based on pyrogenically prepared silicon dioxide.
 - 18. Sweetener comprising granules based on pyrogenically prepared silicon dioxide.
 - 19. Aroma comprising granules based on pyrogenically prepared silicon dioxide.
- 25 20. Feedstuffs additives comprising granules based on pyrogenically prepared silicon dioxide.
 - 21. Chemical intermediates comprising granules based on pyrogenically prepared silicon dioxide.

- 22.Plant protection agents comprising granules based on pyrogenically prepared silicon dioxide.
- 23. Herbicides comprising granules based on pyrogenically prepared silicon dioxide.
- 5 24. Insecticides comprising granules based on pyrogenically prepared silicon dioxide.
 - 25. Fungicides comprising granules based on pyrogenically prepared silicon dioxide.

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A. CLASSI IPC 7	FICATION OF SUBJECT MATTER C01B33/18 B01J20/10 C09C1/3	0	
	o International Patent Classification (IPC) or to both national classific	cation and IPC	
	SEARCHED cumentation searched (classification system followed by classification)	lion symbols)	
IPC 7	CO1B BO1J CO9C		
	·		
Documenta	ion searched other than minimum documentation to the extent that	such documents are included. In the fields so	earched
Electronic d	ata base consulted during the international search (name of data ba	ase and, where practical, search terms used)
EPO-In	ternal	·	•
C. DOCUM	ENTS CONSIDERED TO BE RELEVANT		
Category °	Citation of document, with indication, where appropriate, of the re	levant passages	Relevant to claim No.
X	EP 1 182 168 A (DEGUSSA AG) 27 February 2002 (2002-02-27)		1,3,5-25
Υ	paragraph '0020!; claims		2,4
		erbel ()	
Y	US 3 830 738 A (ARNOLD GEORGE CO ⁻¹ 20 August 1974 (1974-08-20)	IIRELL)	2,4
•	the whole document	٠.	•
v	ED 1 201 722 A (DECHECA)		
T	EP 1 281 733 A (DEGUSSA) 5 February 2003 (2003–02–05)		2,4
;	claims		
x	US 2003/089279 A1 (JURGEN MEYER E	ΕΤ ΔΙ)	1-25
Λ	15 May 2003 (2003-05-15)	- (AL.)	1-25
	the whole document		
		-/	
		,	
		<u></u>	
X Furth	er documents are listed in the continuation of box C.	X Patent family members are listed in	п аппех.
° Special ca	tegories of cited documents :	*T* later document published after the inter	national filing date
	nt defining the general state of the art which is not ered to be of particular relevance	or priority date and not in conflict with to cited to understand the principle or the	
	ocument but published on or after the international	invention "X" document of particular relevance; the cl	atmed invention
"L" docume	nt which may throw doubts on priority claim(s) or	cannot be considered novel or cannot involve an inventive step when the doc	cument is taken alone
citation	or other special reason (as specified) ent referring to an oral disclosure, use, exhibition or	"Y" document of particular relevance; the cl cannot be considered to involve an inv document is combined with one or more	entive step when the
other n		ments, such combination being obvious in the art.	
	an the priority date claimed	*&* document member of the same patent f	amily
Date of the a	ectual completion of the international search	Date of mailing of the international sear	ch report
1:	October 2004	22/10/2004	
Name and m	nailing address of the ISA European Patent Office, P.B. 5818 Patentiagn 2	Authorized officer	
	European Patent Office, P.B. 5818 Patentiaan 2 NL – 2280 HV Rijswijk Tel. (+31–70) 340–2040, Tx. 31 651 epo nl,	D	
	Fax: (+31-70) 340-3016	Bogaerts, M	1

Interioral Application No PCT/EP2004/006719

EP 0 808 880 A (DEGUSSA AG) 26 November 1997 (1997-11-26) the whole document X	C.(Continu	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
16 June 1992 (1992-06-16) abstract "Ullmann's Encyclopedia of Industrial Chemistry - Volume A23" 1993, VCH VERLAGSGESELLSCHAFT , WEINHEIM , XP002300084 page 635 - page 641	X	26 November 1997 (1997-11-26)	1-25
Chemistry - Volume A23" 1993, VCH VERLAGSGESELLSCHAFT, WEINHEIM , XP002300084 page 635 - page 641	X	16 June 1992 (1992-06-16)	
	X	Chemistry - Volume A23" 1993, VCH VERLAGSGESELLSCHAFT , WEINHEIM , XP002300084	1-25
	·		· ·
		4	
	·		
	:.		

Form POT/ISA/210 (continuation of second sheet) (January 2004)

PCT/EP2004/006719

Patent document		Publication	·	Patent family	· · ·	Publication
cited in search report		date		member(s)		date
EP 1182168	A	27-02-2002	EP	1182167	Δ1	27-02-2002
EP 1182168	H	27-02-2002		1186573	–	13-03-2002
			EP		·	
			EP	1182168	·	27-02-2002
			AT	266602	•	15-05-2004
			DE	50102242	_ _	17-06-2004
			JP	2002145610		22-05-2002
			US	2004116270		17-06-2004
			US	2002044903	A1	18-04-2002
US 3830738	Α	20-08-1974	US	3963627		15-06-1976
55 5550755	, ,	20 00 207.	AT	326490		10-12-1975
			AT	132371		15-02-1975
			BE	762674		09-08-1971
						14-05-1974
			CA	947061		
			CH	568242		31-10-1975
•			DE	2107082		26-08-1971
			FR			05-11-1971
			GB	1348372		13-03-1974
			IL	36094	Α	28-07-1975
	•		JP	53028399	В	14-08-1978
			NL .	7102001	Α	18-08-1971
			NO	136074	В	12-04-1977
			SE	401094	_	24-04-1978
			TR	16794		01-05-1973
	,		ne -	10100400		* 2 00 0003
EP 1281733	Α	05-02-2003	DE	10138492		13-02-2003
			BR	0202882	_	03-06-2003
			CN	1405085	A	26-03-2003
			EP	1281733	A1	05-02-2003
			JP	2003160327	A	03-06-2003
			US	2003162881	A1	28-08-2003
US 2003089279	A1	15-05-2003	DE	10123950	A1 ·	28-11-2002
03 2003009279	ΛI	15 05 2005	EP	1266864		18-12-2002
			JP	2003081626		19-03-2003
EP 0808880	Α	26-11-1997	DE	19616781		06-11-1997
			CA	2203726		26-10-1997
			CN	1167729	A,B	17-12-1997
			DE	59709069	D1	13-02-2003
			EP	0808880	A2	26-11-1997
			JP			11-08-2004
			JΡ	10087317		07-04-1998
			KR	260325		01-07-2000
			US	5959005		28-09-1999
			·			
US 5122518	Α	16-06-1992	DE	3835592		26-04-1990
			AT	71801		15-02-1992
			AU	612521		11-07-1991
			ΑU	4292389	Α	26-04-1990
			BR	8905299	Α	22-05-1990
			CA	2000960	_	19-04-1990
			DE			05-03-1992
			EP	0367934		16-05-1990
			ES	2028427		01-07-1992
			GR JP	3004288 2169506		31-03-1993
			. IP	フェムひんけん	A	29-06-1990
			JP	6006522	-	26-01-1994

Interioral Application No PCT/EP2004/006719

KR	9301248 B1	22-02-1993
		FF OF 1770
NZ	231034 A	26-03-1991
TR	24765 A	09-03-1992
ZA	8907473 A	25-07-1990
	TR	TR 24765 A

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